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**PHYSICA B**

## High field study of the magnetic phase transition of URu<sub>2</sub>Si<sub>2</sub>

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### Abstract

We investigate the phase transition of URu<sub>2</sub>Si<sub>2</sub> at  $T_0 = 17.5$  K by thermal expansion and heat capacity in magnetic fields,  $H$ , up to 25 T. At  $T_0$  an energy gap of  $\Delta \simeq 115$  K opens, which decreases to 65 K in 25 T. The  $H$  dependence of  $T_0$  scales with  $\Delta$ , whereas a different  $H$  dependence and energy scale was observed for the ordered dipole moment. The anomalous  $H$  dependence of the thermal expansion coefficients may discriminate between the various scenarios for the primary order parameter of URu<sub>2</sub>Si<sub>2</sub>.

**Keywords:** URu<sub>2</sub>Si<sub>2</sub>; Antiferromagnetism; High magnetic field

The origin of the putative magnetic phase transition [1] in the heavy-fermion superconductor URu<sub>2</sub>Si<sub>2</sub> remains elusive [2,3]. Since the entropy released in the phase transition is very large compared to the antiferromagnetically ordered moments of  $\mu_{\text{ord}} = 0.04\mu_B/\text{U-atom}$  [4], purely dipolar ordering can be ruled out. However, no definitive identification of the primary order parameter has yet been made. It has been suggested that the transition may involve multipolar ordering of uranium ions with a  $5f^2$  configuration, with the implication that the small moment arises from an admixture of higher lying  $5f^1$  or  $5f^3$  states [5,6]. Previously, we have demonstrated [7] by resistivity measurements up to 25 T that the field dependence of  $T_0$  scales directly with the energy gap  $\Delta$ , which opens at  $T_0$ , following  $\Delta/\Delta^{H=0} \propto T_0/T_0^{H=0} \propto 1 - (H/H_0)^2$ . Here the critical fields for  $T_0$  and

$\Delta$ , are equal:  $\mu_0 H_0 = 40 \pm 2$  T. Neutron diffraction [8] indicated a substantial reduction (25%) of  $\mu_{\text{ord}}$  with modest applied fields up to 8 T  $\parallel c$ . The extrapolated critical field for the disappearance of  $\mu_{\text{ord}}$  of  $14.5 \pm 0.3$  T is much smaller than that for  $T_0$  and  $\Delta$ , thereby suggesting that there are two different energy scales relevant to the magnetic phase transition of URu<sub>2</sub>Si<sub>2</sub> [7]. Hence the application of high fields might be the appropriate experimental method towards discriminating between the various scenarios put forth to explain this transition.

Here we extend the field range over which the energy gap can be reliably determined up to 25 T by analyzing the exponential  $T$  dependence of the thermal expansion,  $\alpha$ , along the main  $a$  and  $c$  axes. We also calculate the uniaxial pressure dependence of  $T_0$  at the various applied fields by means of the Ehrenfest relation. The anomalous behavior observed above about 15 T is discussed to stimulate further work in this high-field region of the phase diagram of URu<sub>2</sub>Si<sub>2</sub> [9].

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The experiments have been performed on annealed single crystalline  $\text{URu}_2\text{Si}_2$ . Thermal expansion up to 25 T was measured with a capacitive method, using  $\text{RuO}_2$  and capacitance thermometry. The heat capacity was recorded up to 16 T using a pulse method. The magnetic field was only applied along the easy  $c$  axis. Fig. 1 displays  $\alpha$  parallel to the  $a$  and  $c$  axes, recorded at fields of 0, 10, 14, 18 and 25 T. The  $H = 0$  data are in good agreement with literature [10]. Cooling through  $T_0$ , the  $a$  axis contracts, while the  $c$  axis length increases. Interestingly, and unexpectedly for a regular magnetic ordering, the anomaly grows and remains very sharp upon increasing  $H$ . Below  $T_0$ , the data can be well described by an (dominating) exponential  $T$  dependence, in addition to the linear and cubic terms for the electronic and phonon contributions:

$$\alpha = \alpha_0 + AT + BT^3 + C \exp(-\Delta/T). \quad (1)$$

In Fig. 2 the gap as derived from the volume expansion,  $\alpha_V = \frac{1}{3}(2\alpha_a + \alpha_c)$ , is shown versus  $H$ . The magnitude of  $\Delta^{H=0}$  is the same as obtained from infrared reflectance, vacuum tunneling, nonlinear susceptibility, heat capacity and neutron scattering.  $\Delta$  decreases quadratically with  $H$ , with a critical field  $\mu_0 H_0 = 37 \pm 2$  T, consistent with Ref. [7].

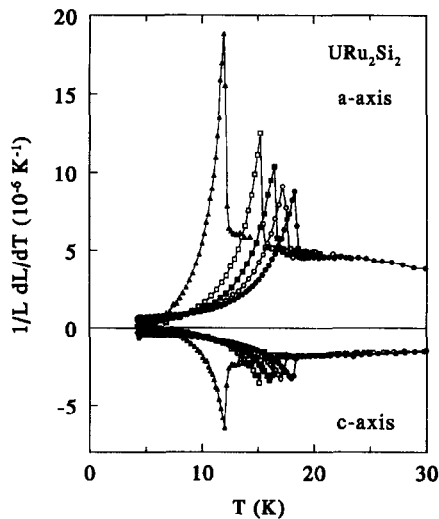


Fig. 1. Thermal expansion of  $\text{URu}_2\text{Si}_2$  along the  $a$  and  $c$  axes, with magnetic fields along the  $c$  axis;  $\mu_0 H = 0$  (●), 10 (○), 14 (■), 18 (□), and 25 T (△).

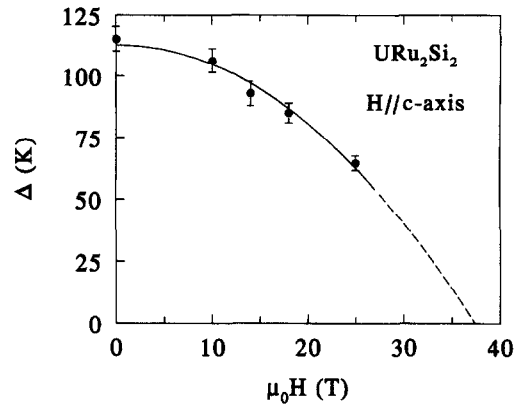


Fig. 2. The energy gap of  $\text{URu}_2\text{Si}_2$  versus magnetic field parallel to the  $c$  axis. The extrapolated critical field equals  $37 \pm 2$  T.

The  $T_0(H)$  phase boundary found in Ref. [7] has been verified by a collection of magnetostriction curves [9]. From the ratio of the jumps at  $T_0$  of thermal expansion and specific heat,  $c$  (not shown), the initial uniaxial pressure dependence can be calculated through the Ehrenfest relation:

$$\left(\frac{\partial T_0}{\partial p_{a,c}}\right)_{B,p'} = \frac{V_m \Delta \alpha_{a,c}}{\Delta(c/T)}. \quad (2)$$

The  $c/T$  jumps at  $T_0$  vary slowly from 0.30 J/mole- $\text{K}^2$  at  $H = 0$  to 0.33 J/mole- $\text{K}^2$  at 16 T [9]. By using  $V_m = 4.95 \times 10^{-5} \text{ m}^3/\text{mole}$ , and assuming that  $\Delta(c/T)$  at 18 T and 25 T does not change significantly from the 16 T value, we can derive the pressure derivatives as shown in Fig. 3. The pressure dependences are almost  $H$ -independent up to about 15 T, but increase strongly at high fields. Note that the anisotropy, reflected in the directly measured  $c/a$  ratio (not shown), changes in a similar way [9], which is a further indication that the above assumption on  $c/T$  is reasonable. The question arises whether this anomalous high-field behavior is due to the diminishing gap, allowing for cross-gap excitations, or perhaps related to the vanishing ordered moment. The observed decrease of  $\gamma = c/T$  at 2 K upon increasing  $H$  [9] makes the first possibility unlikely.

We have presented the first thermal expansion experiments on  $\text{URu}_2\text{Si}_2$  in magnetic fields up to

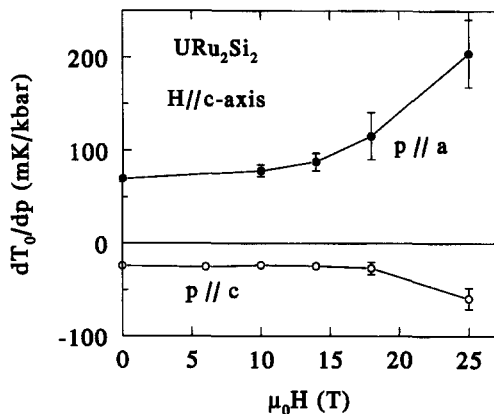


Fig. 3. Initial uniaxial pressure dependence of  $T_0$  for fields up to 25 T, calculated from the Ehrenfest relation. We have assumed that the specific heat jump at 18 and 25 T retains the measured value at 16 T. For this reason, the error bars at these fields are given as twice the experimental uncertainty.

25 T. While confirming the equivalent field dependence of  $T_0$  and  $\Delta$  found in resistivity, the gap derived from  $\alpha_V$  equals that found in tunneling and neutron scattering, and decreases by 45% at 25 T. The  $c/a$  ratio and, possibly also the uniaxial pressure dependence of  $T_0$ , have qualitatively different behaviors in low fields ( $\mu_0 H < 15$  T) and high fields ( $\mu_0 H > 15$  T). Such behavior would be consistent with the presence of an extra energy scale of about

15 T, proposed to relate to the disappearance of the staggered moment. More experiments in this field range are necessary to substantiate such speculations and could eventually enable us to identify the primary order parameter of  $\text{URu}_2\text{Si}_2$ .

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